What is claimed:

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1. A method of using silyl chemistry to control the reactivity of a self-assembled molecular electro-optic material, said method comprising:

providing an electro-optic material comprising a silyl-derivatized chromophore;

desilylating said chromophore to generate terminal hydroxy functionalities; and

reacting said hydroxy functionalities with a reagent having at least one silicon moiety.

2. The method of Claim 1 wherein said chromophore is a high-β chromophore.

3. The method of Claim 1 wherein said chromophore is derivatized with a trialkylsilyl protecting group.

4. The method of Claim 3 wherein said chromophore is derivatized with a *tert*-butyldimethylsilyl protecting group.

5. The method of Claim 1 wherein said chromophore is desilylated by treatment with a deprotecting agent.

6. The method of Claim 1 wherein said chromophore is derivatized with a *tert*-butyldimethylsilyl protecting group.

7. The method of Claim 6 wherein said chromophore compound is desilylated with a quaternary ammonium fluoride.

8. A method of using silyl chemistry to generate a hydrophilic surface for molecular self-assembly of an electro-optic material, said method comprising:

providing an electro-optic material comprising a high-β chromophore film with terminal trialkylsiloxy moieties;

desilylating said film to generate terminal hydroxy functional groups; and

reacting said terminal hydroxy functional groups with a siloxane capping agent.

9. The method of Claim 8 wherein said film is desilylated by nucleophilic reaction at trialkylsiloxy moieties.

10. The method of Claim 8 wherein said film has terminal *tert*-butyldimethylsiloxy moieties.

11. The method of Claim 10 wherein said film is desilylated with tetra-*n*-butylammonium fluoride.

12. A method for assembling a multi-layered electro-optic siloxane film, said method comprising:

providing a substrate with a hydroxylated surface;

coupling a chromophore layer to said surface, said layer comprising a plurality of chromophore molecules, each said molecule reactive with said surface and having a terminal trialkylsiloxy moiety;

desilylating said chromophore layer to generate terminal hydroxy functionalities; and

coupling said chromophore layer with a capping layer, said capping layer comprising molecular components and each said component having at least two silicon moieties, said coupling providing a siloxane bond sequence between said chromophore and capping layers.

13. The method of Claim 12 wherein said chromophore molecule is a high-β chromophore.

14. The method of Claim 13 wherein each said high-β chromophore has a terminal *tert*-butyldimethylsiloxy moiety.

15. The method of Claim 12 wherein said chromophore is desilylated by reaction of a nucleophile with said trialkylsiloxy moiety.

16. The method of Claim 15 wherein each said high-β chromophore has a terminal *tert*-butyldimethylsiloxy moiety.

17. The method of Claim 16 wherein said chromophore is desilylated with tetra-*n*-butylammonium fluoride.

18. The method of Claim 12 wherein said capping layer comprises octachlorotrisiloxane.

19. The method of Claim 18 wherein a second chromophore layer is coupled to said capping layer, said second chromophore layer comprising a plurality of chromophore molecules, each said chromophore molecule reactive with said capping layer and having a terminal trialkylsiloxy moiety.

20. The method of Claim 18 wherein said second coupled chromophore layer is desilylated then coupled with a second capping layer.

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21. A non-linear optical article comprising a plurality of molecular bilayers, each said bilayer comprising a first chromophore molecular layer coupled to a capping molecular layer with a siloxane bond sequence, said capping molecular layer capable of coupling to another chromophore molecular layer with a siloxane bond sequence.

22. The material of Claim 21 wherein said chromophore is a high-β chromophore.

23. The material of Claim 21 wherein said capping layer is a polysiloxane.

24. The material of Claim 23 wherein said capping layer comprises octachlorosiloxane.

25. The material of Claim 21 wherein said bilayers are deposited on a substrate.

26. The material of Claim 25 wherein said substrate and said bilayers are incorporated into a waveguide device.

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27. A chromophore composition with non-linear optical properties having the structural formula $(Ch)XR_n$, wherein (Ch)X is a chromophore substructure and X is a heteroatom; R is a trialkylsiloxyalkyl moiety; and n is the number of said moieties meeting the valence requirement of said heteroatom.

28. The composition of Claim 27 wherein said chromophore is selected from the group consisting of structural formulae shown in FIGS. 2,11 and 15.

29. The composition of Claim 27 where in X is selected from the group of heteroatoms consisting of O and N.

30. The composition of Claim 29 wherein X is N and n is 2.

31. The composition of Claim 27 comprising a non-linear optical film.